DETCHATION OF FAMILIES DISPERSED POWDERS IN AIR

HAVID C. HEBERLEIN, PhD US ARMY MOBILITY EQUIPMENT RED COMMAND FURT BULNOIR, VIRGINIA

## INTRODUCTION

The future battlefield will have a high density of sophisticated, lethal weapons. Mines will be used on this battlefield together with nuclear, biological and chemical weapons to retard and restrict the advance of enemy forces across wide land areas. A wide variety of remotely deliverable, air scatterable mines will be available to interdict, delay and destroy enemy forces. For US forces to remain effective in this hostile environment, friendly forces must be able to maintain their tempo of operations under fire. These unique requirements dictate the development of real time countermine systems, effective against a wide variety of fuze types, that quickly clear large land areas. Explosive technology is one of a small number of technological areas capable of meeting these special mission requirements.

Traditionally, explosive weapons such as line charges have been used to produce single impulses sufficient to deflect mine pressure plates and thereby cause mine detonation. Recent advances in the development of fuel air explosive (TAE) weapons has lead to mine neutralization systems that are effective against pressure plate mines over wide land areas. Present FAE weapons explosively disseminate and detonate a liquid fuel over a wide land area. A small central explosive burster disperses the liquid fuel as small droplets suspended in a large air mass. A detonator, also dispensed into the fuel-air cloud, is detonated after an appropriate delay. While these liquid fuel systems work well for mine neutralization applications, there are shortcomings to this approach: (1) the maximum pressures obtainable for stoichiometric combustion; (2) approximately 10% of the liquid fuel rounds result in burning and not in detonation; (3) long term storage of certain fuels results in polymerization of the liquid. The use of solid powders in wide area mine neutralization weapons obviates many of the difficulties

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intrinsic to liquid fuels. Solid particle sizes can be controlled during manufacture so that explosive dispersal can be accurately and reliably controlled. In addition, solids have long shelf lives reducing the problems associated with long term storage of munitions. Most importantly, however, the terminal weapon effects for liquid fuel weapons are limited by the pressures they can produce.

The vulnerability of a structure to blast overpressures is given by a critical load curve in which peak pressures are plotted versus impulse. The changing values of Y, I given by each curve is shown schematically in Figure 1 together with an indication of the performance of FAE weapons against these mines. The detonation of dispersed powders does not share the limitation in maximum overpressures found for the liquid fuel-air systems. Solid powders such as explosive powders and combinations of flaked aluminum and ammonium perchlorate, carry their own oxygen at the molecular level for efficient combustion during detonation.

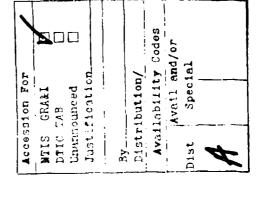
The pressure P, obtainable in detonation of a powder-air mixture is a function of the heat in a particular solid available to be released in detonation, Q, the density of explosive, P, and the detonation velocity, D, such that

$$P = P(\rho, Q, D). \tag{1}$$

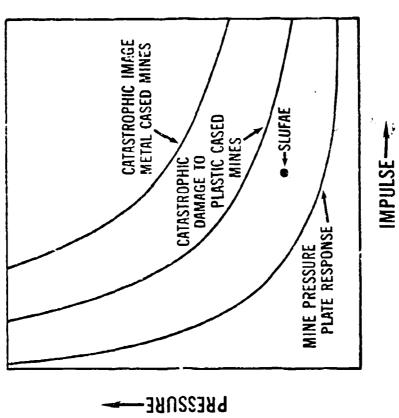
The maximum pressures attainable in powdered, dispersed solid systems that carry molecular oxygen is easily related to the density of explosive, the energy available for combustion and the detonation velocity. Therefore, dispersed powdered systems can be developed in which the pressures obtained are determined by the density of the dispersed powder-air mixture and the combustion energy intrinsic to the selected solid.

### THEORETICAL BASIS FOR THE DEFONATION OF POWDERS DISEERSIND IN AIR

A theoretical model of the detonation of fine powders dispersed in air must accurately describe the exothermic release of chemical energy, the compressibility of the reaction products and chemical reaction rate effects. If it can be assumed that the reaction products are gaseous and rapidly mixed and that reaction rate effects are manifested in a detonation wave propagating through the unreacted mixture of a powder suspended in air, then details of the detonation process at the particle scale will not substantially affect detonation parameters. The dispersed powder-air mixture can be represented as a molecular, heterogeneous medium at an averaged thermodynamic state. The Chapman-Jouguet (CJ) detonation wave model is appropriate for this medium if the reaction







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FIGURE 1. LAND MINE STRUCTURE YIELD CURVES

front is handled as a discontinuity across which an average instantaneous energy is released. Flickett and Davil have shown from energy, mass and momentum relations at the CJ discontinuity that

$$P_{o} = 2(\chi - 1) f_{\infty} E, \qquad (2)$$

$$f_0 = \frac{K+1}{K} f_{\infty}, \qquad (3)$$

$$D = \left[ \frac{(K+1)P_0}{f_{\infty}} \right]^{1/2}, \text{ and}$$

$$f_{\infty} = f_{\text{air}} + \left\{ 1 - \frac{f_{\text{air}}}{f_{\text{out}}} \right\} \in \mathcal{L},$$

$$f_{\text{is the detonation pressure.}}$$

$$K \text{ the ratio of the specific heat}$$

where Po is the detonation pressure, the ratio of the specific heats of the combustion products. To the loading density, E the exothermic reaction energy, the density and the CJ state, D the detonation velocity and C the concentration.

The amount of energy released depends on the chemical and thermodynamic processes involved in detonation. A fundamental assumption in the Chanman-Jouguet theory of detonation is that chemical equilibrium exists among the reaction species in the reaction zone of the detonation, i.e. all of the chemical reaction energy contributes to the propagation of the detonation. It is convenient to evaluate the energy, E, for a constant volume adiabatic process so that the result can be interpreted as independent of such combustion properties as the nature and distribution of solid particles. The constant volume pressure, Pv, is given by

$$P_{\nu} = (N-1) \cdot \rho_{\infty} E. \qquad (6)$$

If the values for K and E are approximately the same  $^2$ , then

$$P_{o} = 2P_{v}. \tag{7}$$

The constant volume pressure, Pv, is easily observed experimentally as the pressure observed for a detonation in a fixed volume "closed bomb".

<sup>&</sup>lt;sup>1</sup>Flickett, W. D., "Detonation", University of California Fress, Berkeley,

<sup>&</sup>lt;sup>2</sup>Zeldovitch, I. B., and Dompaneets, A. S., "Theory of Detonation", Academic Press, New York, 1900.

To compare this experimental value for the pressure given in equation 7 with that theoretically expected for a specific powder, the theoretical pressure is obtained for the temperature of the detonation. The temperature attained depends on the heat evolved in chemical reactions less that consumed in physical phase changes and the heat capacifies of the reaction products obtained. The mathematical statement for chemical equilibrium of multi-reaction is a minimum. The Gibbs free energy is minimized using a La Grange multiplier technique developed by Naphtali<sup>3</sup>. A table of stoichiometric coefficients is formed in which each column represents a reaction and each row a component. The energy in row i column is

$$y_{ij} = (\partial \pi_i)/(\partial \xi_i), \qquad (8)$$

where  $\pi_i$  is the number of moles of component i and  $\xi_i$  is the extent of reaction i. A change in composition is given by

$$d\pi_{i} = \sum_{j} \nu_{ij} d\xi_{j} . \tag{9}$$

According to thermodynamics,  $G = \sum \mu_i \pi_i$  where  $\mu_i$  is the standard free energy of formation per mole. At a fixed temperature and pressure, we obtain

$$dG = \sum_{i} \mu_{i} \left\{ \sum_{j} \nu_{ij} d\xi_{j} \right\} = \sum_{j} \left\{ \sum_{i} \mu_{i} \nu_{ij} \right\} d\xi_{\cdot (10)}$$

The free energy change for reaction j can be defined as

$$\Delta G_{j} = \sum_{i} \mu_{i} \gamma_{ij}, \qquad (11)$$

so that equation 10 becomes

Naphtali, N. M.. "Calculate Complex Chemical Equilbira", Ind. Engr. Chem., 53, 387, 1961

$$dG = \sum_{j} \Delta G_{j} d\xi_{j} . \tag{12}$$

For equilibrium dG=0 requires that  $\Delta G_j=0$ . The condition  $\Delta G_j$  forms a set of non-linear, equilibrium equations. The difficulty in minimizing  $\Delta G_j=0$  increases as the number of equations increases.

Any change in composition is thermodynamically possible if  $dG \le 0$  A series of these changes should lead to a minimum point, i.e. equilibrium composition. By choosing  $d\xi_1 = -\Delta G_1 d\lambda$  for  $d\lambda \ge 0$ ,

$$dG = -\left\{\sum_{i} (\Delta G_{i})^{2}\right\} d\lambda. \qquad (13)$$

A change in composition is then given by

$$dn_{i} = -\left\{\sum_{j} y_{ij} \Delta G_{j}\right\} d\lambda = -\sum_{k} \left\{\sum_{j} y_{ij} y_{kj}^{k}\right\} d\lambda.$$
(14)

A symmetric table of values  $\mathcal{E}_{i \bullet}$  can be formed for the condition

$$\mathcal{E}_{ik} = \sum_{j} \nu_{ij} \nu_{kj} = \mathcal{E}_{ki}, \qquad (15)$$

so that

$$\frac{(d\pi_i)}{(d\lambda)} = -\sum_{k} E_{ik} \mu_k \tag{16}$$

gives the direction of decreasing free energy for any composition. Using Equation 10

$$\frac{(d6)}{(d\lambda)} = \sum_{\mu_i} \left[ \frac{(dn_i)}{(d\lambda)} \right] = -\sum_{j} (\Delta G_j)^2, \quad (17)$$

the proximity to equilibrium can be obtained from the sum of the square for the  $\Delta G_j$ 's for all reactions. Near equilibrium each  $\Delta G_j$  should be near zero and also the sum,  $\sum_i (\Delta G_j)^2$ .

For the detonation of aluminum powders in air, the potential reactants and products include the following.

#### TABLE I

REACTANT	PRODUCT		
Al(s) Al(s), Al(1), Al(g)			
$Al_20_3(s)$ $Al_20_3(s)$ , $Al_20_3(1)$ $Al_20_3(g)$			
$N_2(g)$ Al0(g), Al02(g), Al2			
C <sub>2</sub> (g)	HA102		
H <sub>2</sub> O(g)	$N_2(g)$ , $O_3(g)$ , $N_3(g)$ , $O(g)$ ,		

Using this set of reactants and products together with thermodynamic heat capacity data for each of these species, chemical equilibrium compositions were calculated as a function of aluminum concentration in air. The result of these calculations for the Chapman-Jouguet Pressure, Equation 7, for aluminum powder detonated in air is shown as a function of aluminum concentration in Figure 2.

## EXPERIMENTAL APPARATUS

The shock tube is a good diagnostic apparatus for studying onedimensional explosive phenomena. Shock tubes can be used to investigate extrinsic properties of powder-air systems such as pressure, temperature and concentration as well as intrinsic properties such as particle size and ignition induction time. Homogeneous powder-air systems were investigated with a detonation tube with the provision for the rapid formation of powder-air mixtures. To assess the powder distribution homogeneity, the concentration of the powder-air mixture was measured through laser

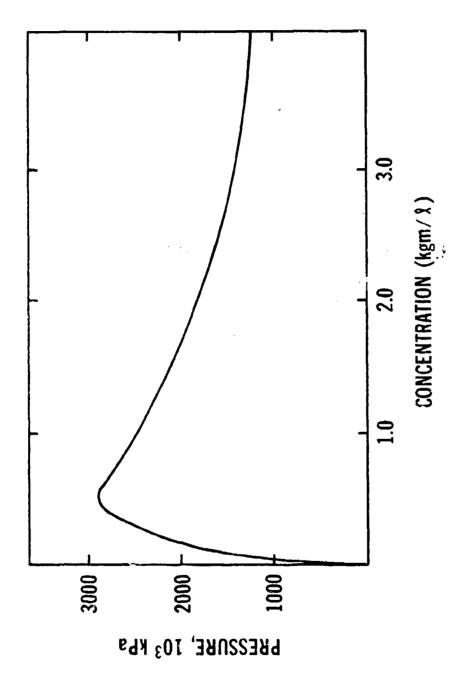


FIGURE 2. CALCULATED CHAPMAN-JOUGUET PRESSURE AS A FUNCTION OF ALUMINUM CONCENTRATION IN AIR

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beam attenuation.

The detention tube condicted of three separate sections, each 1.8m in length. The chook tube year mounted vertically for a total height of 5.4m. Each steel tube rection had an internal diameter of 152mm and a 25mm wall thickness. The chook tube was designed for static pressures of 14.8GPa. Special clamps were used to hold the three sections of the tube together. Both ends of the detonation tube were open. Test powder was introduced into the bottom and vented at the top of the tube. A blower was placed at one end of the tube to pass air through the tube continuously. A sunnometer was used to monitor the air flow through the shock tube. The entire shock tube assembly is shown schematically in Figure 3.

Pressure measurements were obtained from piezoelectric gages mounted in the center test section. These gages were mounted in Nylon-6 to dampen acceleration effects from the shock tube. A thin coating of silicone grease containing powdered graphite was used to prevent a thermal response from the gages. All gages were initially calibrated and routinely recalibrated in situ using known shock pressures. Pressure gage responses were amplified by Tektronic AM502 amplifiers and recorded on a Bell and Howell 3700E tape recorder. The concentration of the powder was monitored by measuring the attenuation of the He-Ne laser (632mm) from a Spectra-Physics Model 132 laser. The attenuated beam was measured by a Hewlett Packard Photodiode, Model 5082-4220.

The detonation tube was calibrated using homogeneous gas phase detonations. Calibration tests used near stoichiometric ethylene-air gas phase mixtures. These tests resulted in detonation velocities of 1859 ± 15 m/sec and a detonation pressure of 298 ± 200 psi, consistent with reported Champan-Jougnet values of 1750 m/sec and 270 psi respectively.

# EXPERIMENTAL RESULTS

The detonation tule was used to assess the deflagration and detonation characteristics of various fuel powders. The results of these tests are shown in Table 2.

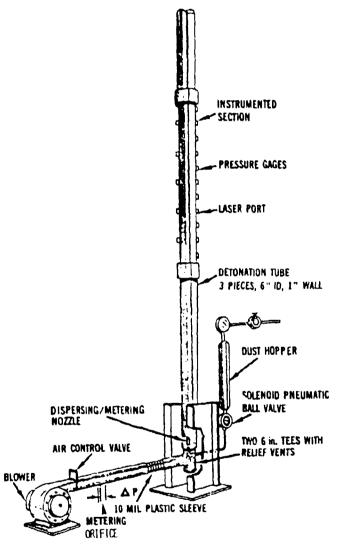


Figure 3. SCHEMATIC DRAWING OF SHOCK TUBE APPARATUS.

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-TABLE 11

	Concentration			
kg/m <sup>3</sup>		13	Propagation Vel	Overpressure
wder	Stoich	Exp	(km/sec)	(kľa)
al	.10	. 36	.5840.02	371
crotheneR	.12	.19	.58±0.04	392
arch	.12	.46	.63±0.03	427
gnesium	.45	.38	.61±0.06	434
uminum	.33	.40	1.54±0.11	5097
uminum	.33	.60	1.62±0.12	6352
uminum	.33	.50	1.46±0.09	5759
uminum	.33	80	1.50±0.10	5434
crotheneR arch gnesium uminum uminum uminum	.10 .12 .12 .45 .33 .33	.36 .19 .46 .38 .40 .60	.58±0.02 .58±0.04 .63±0.03 .61±0.06 1.54±0.11 1.62±0.12	371 392 427 434 5097 6352 5759

All of these powder dispersions propagated a shock wave disturbance based on film records and pressure measurements. However, events involving coal dust, Microthene<sup>8</sup>, starch and magnesium were considered to be deflagrations. The flaked aluminum powder did detonate. Sedgwick et. al. gives the variation of the detonation velocity with concentration of aluminum in air shown in Figure 4. It is possible that the variance of the experimental data (approximately 1.6km/sec) from the Chapman-Jouguet velocity (1.8km/sec) was due to variation from stoichiometric concentration, although wall losses due to inadequate tube size could also account for a decreased detonation velocity. The extended reaction zone found in two phase heterogeneous systems (aluminum in air) also contributes to differences in experimental data from that calculated for Chapman-Jouguet conditions. Sichel and Schaaff<sup>5</sup> have shown that it is

Sedgwick, R. T., Kratz, H. R. and Baker, M., "Concepts for Improved Fuel Air Explosives", Systems, Science and Software Report SSR-R-77-3005, September 1976.

Sichel, M. and Schaaff, J., "Excl. Air Explosives - Theoretical Considerations", AFATL-TR-71-171, 1, 109, December 1971.

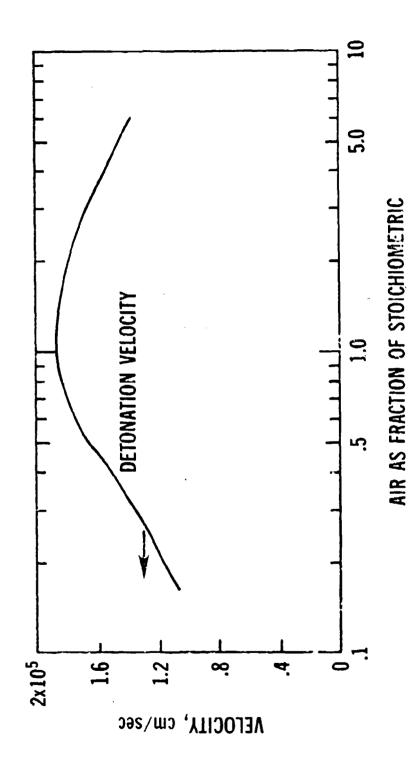


FIGURE 4. CALCULATED DETONATION VELOCITIES FOR CHAPMAN-JOUGUET DETONATION IN ALUMINUM-AIR MIXTURES AS A FUNCTION OF CONCENTRATION

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necessary to consider the fact that fuel particles do not contribute to the pressure or volume occupied by the unreacted fuel (powder) and air mixture. When this aspect of two-phase detonations is considered, then it has been shown that the detonation velocity of the two-phase system is lower and the detonation pressure higher than that which could be calculated for a purely gaseous detonation.<sup>6</sup>, 7

Figure 2 gives the calculated Chapman-Jouguet pressures for the detonation of various aluminum concentrations in air. The calculated Chapman-Jouguet pressure neithere exceeds 3,000 kPa, yet stable pressures were observed experimentally up to twice this value. The experimental pressure responses are believed to be accurate to the degree indicated in Table II. Gidaspow<sup>8</sup> has pointed out that in two-phase flow there are also complex effects that can cause instability effects that may be manifested in the pressure and other characteristics of heterogeneous detonations. It has been demonstrated that disproportionation exists in the induction zone of propagating two-phase detonations. This implies unequal phase velocities of the two phases and a more complex analysis of the instability effects. Alternatively spinning detonation of twophase systems also increases the observed pressure while lowering the detonation tube at a lower linear velocity (and a higher rotational velocity) and creates substantially higher pressures at the walls of the detonation tube.

Williams, F. A., "Structure of Petonations In Dilute Sprays", Phys. Fluids, 4, 1961.

Nicholls, J. A., Debora, E. K. and Ragland, K. W., "A Study of Two-Phase Detonation As It Relates To Rocket Metor Combustion Instability", NASA CR 272, 1965.

<sup>&</sup>lt;sup>8</sup>Gidaspow, D., "A Thermodynamic Theory of Two-Phase Flow With Unequal Phase Velocities", NATO Advanced Study Institute on Two-Phase Flows and Heat Transfer, August 1976.

#### CONCLUSIONS

Fine powders of flaked aluminum have been rapidly dispersed and then detonated in air. These powder-air mixtures produce substantial overpressures. Chemical combustion reactants and products have been identified for an aluminum detonation in air. A La Grange multiplier technique was used to minimize the Gibbs Free energy associated with the chemical equilibrium combustion products. Chapman-Jouguet pressures and detonation velocities were calculated for the combustion of aluminum powders in air over a large range of powder concentrations.

Experimentally, a fluidized shock tube was constructed to study the detonation of heterogeneous powder-air systems over a wide range of cloud densities. Dynamic particle concentrations were measured as a function of cloud overpressure and detonation velocity. Experimental measurements of pressures, detonation velocities and cloud densities indicated that dispersed powders of coal dust, MicrotheneR, starch, magnesium and aluminum propagated shock wave disturbances. The events involving coal dust, MicrotheneR, starch and magnesium were determined to be deflagrations. The flaked aluminum powder detonated over a wide range of cloud densities. The aluminum cloud pressures observed experimentally were higher than those reported for liquid fuel air explosive clouds. The pressures observed for the abminum-air of louds were higher than those calculated by minimizing the Gibbs Free energy. Correspondingly, lower detonation velocities were obscrved than those obtained from theoretical calculations. These variances are attributed to the extended reaction zone found in two-phase, heterogeneous systems.

The detonation of dispersed powders has a great potential for future weapons to generate significantly higher pressures over wide land areas than that presently available through present liquid fuel air explosives. Because powders (explosives, combinations of fuel and exidizer such as aluminum plus teflom or ammonium perchlorate, etc.) carry their own oxygen at the molecular level, the pressures generated by the detonation of these powder-air clouds will not be restricted by the amount of atmospheric oxyger available for combustion. In fact, the pressures generated by these clouds can be controlled through the density of the powder-air clouds and the chemical energy intrinsic to the solid powders used to generate each cloud. The higher pressures and impulses from these clouds will be capable of causing catastrophic damage to land mimes or other structures on the battlefield. Used in a countermine role, the development of powder-air systems represents a potential for significant new capability to destroy or neutralize mines over wide land areas irrespective of mine fuzing.